**NDAG TASKING 2007-01**
Date Issued: 2/06/07

Task Title: Technical Peer Review of ORNL U233 Dilution Report

Task Statement: The NDAG is requested to provide an independent technical peer review of the attached ORNL U233 Dilution Report. Please pay particular attention to the final dilution ratios as they will be used in support of the ORNL Building 3019 U233 processing mission.

Task Deliverable: Written (transmission by email is acceptable) report.

Task Due: April 27, 2007

**Response to NDAG TASKING 2007-01**
April 16, 2007

**Technical Peer Reviews of ORNL/TM-13524: "Isotopic Dilution Requirements for $^{233}U$ Criticality Safety in Processing and Disposal Facilities"**

## Overview

Review comments from three independent reviewers are attached. One reviewer focused primarily on the recommended dilution ratios; the other two provided more general reviews.

General comments shared by these reviews included:

- The approach to achieve criticality safety in processing and disposal of $^{233}U$ via dilution with depleted uranium (DU) is well justified.
- Limitation of the scope of the study to U mixtures with silicon dioxide ($SiO_2$) and water covers the most reasonable range of mixtures in disposal scenarios but does not consider likelihood of other limiting cases with other moderators.
- Selection of the subcritical value for the infinite-media neutron multiplication factor ($k_\infty$) for the $^{233}U$ mixtures as $\leq 0.95$ (later stated as "not fully justified" by the authors) was considered arbitrarily conservative.
- Consideration of heterogeneous environments (as in analysis of a repository) was beyond the scope of this study. Analysis of homogeneous infinite media without absorbers maximizes conservatism and cost.
- Methods and data for the analyses are reasonable, though dated at the time of the study (1997) and much more dated today. It is recommend that select maximum $k_\infty$ values be verified using modern methods and data. For example, isotopic evaluations are available for Si in ENDF/B-VII.0 versus the elemental Si evaluation in ENDF/B-V.

Reviewers verified the expressions for the dilution ratios for the fast (un-moderated) and thermal (moderated) homogeneous binary/ternary (U233/HEU/DU) mixtures. The study asserts that no combinations of U mixtures with silicon dioxide and water were more reactive than "The limiting subcritical enrichment for $^{235}U$ (Paxton and Pruvost, July 1977) for optimally moderated homogeneous aqueous systems is well-defined to be 1 wt % $^{235}U$" – leading to the use of that criteria for all moderator mixtures. This premise initiated reviewer questions regarding other limiting cases with these moderators and with other moderators.

Future application of this report should consider the specific comments and questions of the individual reviews.

Finally it was noted that development of an NCSET module on hand calculation of dilution ratios would be of value.

**Review A**

## Fast Dilution Ratio

Table 1. Unmoderated Infinite Homogeneous "Fast" Media

| Parts per Atom | | | | | | k-infinity | | COG |
|---|---|---|---|---|---|---|---|---|
| $^{232}U$ | $^{233}U$ | $^{234}U$ | $^{235}U$ | $^{236}U$ | $^{238}U$ | Stewart | Leal | File/s |
| *Binary (and Ternary) Dilution Ratios (with DU)* | | | | | | | | |
| | 3.25 | | | | 96.75 | 0.998(1) | 0.992(1) | u325/u325r |
| | 3.1 | | 0.2 | | 96.70 | 0.992(1) | | u320 |
| | | 30.0 | | | 70.00 | 0.998(1) | | u234 |
| | | 29.0 | 0.142 | | 70.858 | 0.999(2) | | u234x |
| | | | 5.56 | | 94.44 | Critical Experiment | | Scherzo 556 |
| | | | 3.3 | 96.7 | | 0.997(1) | | u236 |
| *Ternary Dilution Ratios* | | | | | | | | |
| | 3.25 | | 0.0 | | 96.75 | 0.998(1) | 0.992(1) | u325/u325r |
| | 3.1 | | 0.2 | | 96.70 | 0.992(1) | | u320 |
| | 2.5 | | 1.25 | | 96.25 | 0.997(1) | | fast1 |
| | 2.0 | | 2.1 | | 95.90 | 0.998(1) | | fast2 |
| | 1.5 | | 2.95 | | 95.55 | 0.997(2) | | fast3 |
| | 1.0 | | 3.8 | | 95.2 | 0.998(2) | | fast4 |
| | 0.5 | | 4.65 | | 94.85 | 0.999(2) | | fast5 |
| | 0.0 | | 5.56 | | 94.44 | Critical Experiment | | Scherzo 556 |

The critical fast dilution constants are then:

DU(0.2)/235U = (94.64/5.36)(238/235) = 17.9 g/g
DU(0.2)/233U = (96.90/3.10)(238/233) = 31.9 g/g

The last result can be compared to the subcritical ($k_\infty$ = 0.95) dilution ratio of gDU/g233U = 35.364 given as 'Result No. 83' given in Table A.1 on page A-11 of ORNL/TM-13524 and rounded to 36 in equation (A.10) on page A-15.

The critical dilution ratio for a mixture of $^{233}U$, $^{235}U$ and $^{238}U$ can be *estimated* by combining these results linearly:

$$31.9(g233U) + 17.9(g235U) = gDU$$

This result is consistent with the (subcritical) fast dilution law given as equation (A.10).

## Thermal Dilution Ratio (Homogeneous)

Table 2. Optimally Moderated Infinite HOMOGENEOUS "Thermal" Media

| Atom Fractions | | | | | | | | k-infinity | | COG |
|---|---|---|---|---|---|---|---|---|---|---|
| $^{233}U$ | $^{234}U$ | $^{235}U$ | $^{236}U$ | $^{238}U$ | H | O | H/X | Stewart | Leal | File/s |
| | | | | | | | | | | |
| 1.01 | | | | 98.99 | 101.0 | 50.5 | 100 | 0.999(2) | | k100 |
| 0.85 | | | | 99.15 | 170.0 | 85.0 | 200 | 1.001(2) | | k200 |
| 0.80 | | | | 99.20 | 240.0 | 120.0 | 300 | 0.997(2) | 0.982(2) | k300/r |
| 0.79 | | | | 99.21 | 316.0 | 158.0 | 400 | 0.998(2) | 0.976(2) | k400/r |
| 0.81 | | | | 99.19 | 405.0 | 202.5 | 500 | 0.997(2) | 0.983(2) | k500/r |
| | | | | | | | | | | |
| 0.82 | | | | 99.18 | 328.0 | 164.0 | 400 | | 0.997(2) | j400r |
| | | | | | | | | | | |
| | | | | | | | | | | |
| | | | | | | | | | | |
| | | | | | | | | | | |
| 0.79 | | 0.0 | | 99.21 | 316.0 | 158.0 | 400 | 0.998(2) | | k400 |
| 0.63 | | 0.2 | | 99.17 | 332.0 | 166.0 | 400 | 0.994(2) | | o400 |
| 0.5 | | 0.37 | | 99.13 | 348.0 | 174.0 | 400 | 0.996(2) | | b400 |
| 0.2 | | 0.75 | | 99.05 | 380.0 | 190.0 | 400 | 0.996(2) | | c400 |
| 0.0 | | 1.0 | | 99.00 | 400.0 | 200.0 | 400 | 0.997(2) | | a400 |
| | | | | | | | | | | |

The (critical) homogeneous dilution ratios are:

233U/238U = 0.79 (Calculation; Stewart 1978)
233U/238U = 0.83 (Calculation; Leal 2001)
235U/238U = 1.0 (PCTR Measurement)

The (critical) dilution ratio for a mixture of $^{233}U$ and DU(0.2) may be estimated from binary data as:

0.79 - 0.2(0.79/1) = 0.63 (from calculation; Stewart 1978); or
0.83 - 0.2(0.83/1) = 0.66 (from calculation; Leal 2001),

which are in excellent agreement with ternary calculations (cases o400 and p400r). Note that ANSI/ANS-8.1 gives a subcritical limit of U(0.93). So corresponding subcritical dilution ratios (for $k_\infty = 0.98$) are:

233U/DU(0.2) = 0.59 (Stewart 1978)
233U/DU(0.2) = 0.62 (Leal 2001)
233U/238U = 0.74 (Stewart 1978)
233U/238U = 0.78 (Leal 2001)
235U/238U = 0.94 (ANSI/ANS-8.1)

which is in good agreement with the ORNL result (for $k_\infty = 0.95$) of:

233U/DU(0.2) = (1/187.6792)(238/233) = 0.54 g/g*
233U/DU(0.2) = 1/188= 0.53 g/g**

*See footnote "a" on page A-8. **See equation (A.11) on page A-15.

**Thermal Dilution Ratio (Heterogenous)**

Minimum (critical) dilution ratios can be estimated for HETEROGENEOUS lattices of uranium metal rods in water based on data in CP-2842 (since optimum moderation occurs at nearly the same H/X (atomic) or $H_2O/U$ (volume) ratio. COG calculations determine the equivalent $^{233}U$ "enrichment" based on the $^{235}U$ data from CP-2842.

The (minimum critical) dilution ratio is 233U/238U = 0.56 (by atom) corresponding to an "enrichment" of 0.55 g(233U)/gU. Considerations of heterogeneity are beyond the scope of the ORNL report. The agreement with the ORNL value of 0.53 wt. % $^{233}U$ (which appears on the bottom of page A-15) is coincidental.

Table 3. Near Optimum Moderated Infinite HETEROGENOUS "Thermal" Media

| Metal | Diam. | Air Gap | $H_2O/U$ | Temp. | $k_{eff}$ | File | Comment |
|---|---|---|---|---|---|---|---|
| | | | | | | | |
| 235U(0.75) | 3.00 cm | None | 1.36 | 20 C | 1.000(1) | U75 | Estimated* |
| 233U(0.57) | 3.00 cm | None | 1.36 | 20 C | 0.999(3) | U57 | This work |
| | | | | | | | |
| 235U(0.71) | 1.10 in | 0.45 in | 1.56 | 25 C | 0.997(1) | TU1 | +Buckling* |
| 233U(0.55) | 1.10 in | 0.45 in | 1.56 | 25 C | 0.997(3) | TU23 | This work |

*CP-2842

## Acceptable Minimum Safety Margins

The $k_{eff} < 0.95$ subcritical limit for this type of application (based on infinite media calculations) has been criticized as unnecessarily restrictive. See "Acceptable Safety Margins and the Multiplication Factor" distributed as a handout to a course in *Criticality Safety Fundamentals with a Focus on NTS Activities and A Review of Worldwide Criticality Accident Statistics with a Focus on Lessons Learned* to NVO and Contractor Personnel by Thomas P. McLaughlin (Los Alamos National Laboratory) on April 4, 2000. The reviewers concur with the position stated in this 'white paper'.

## Hand Calculations

Determination of dilution ratios could be developed into an NCSET module to demonstrate the use of integral (spectrum weighted) cross-sections with simple hand calculations.

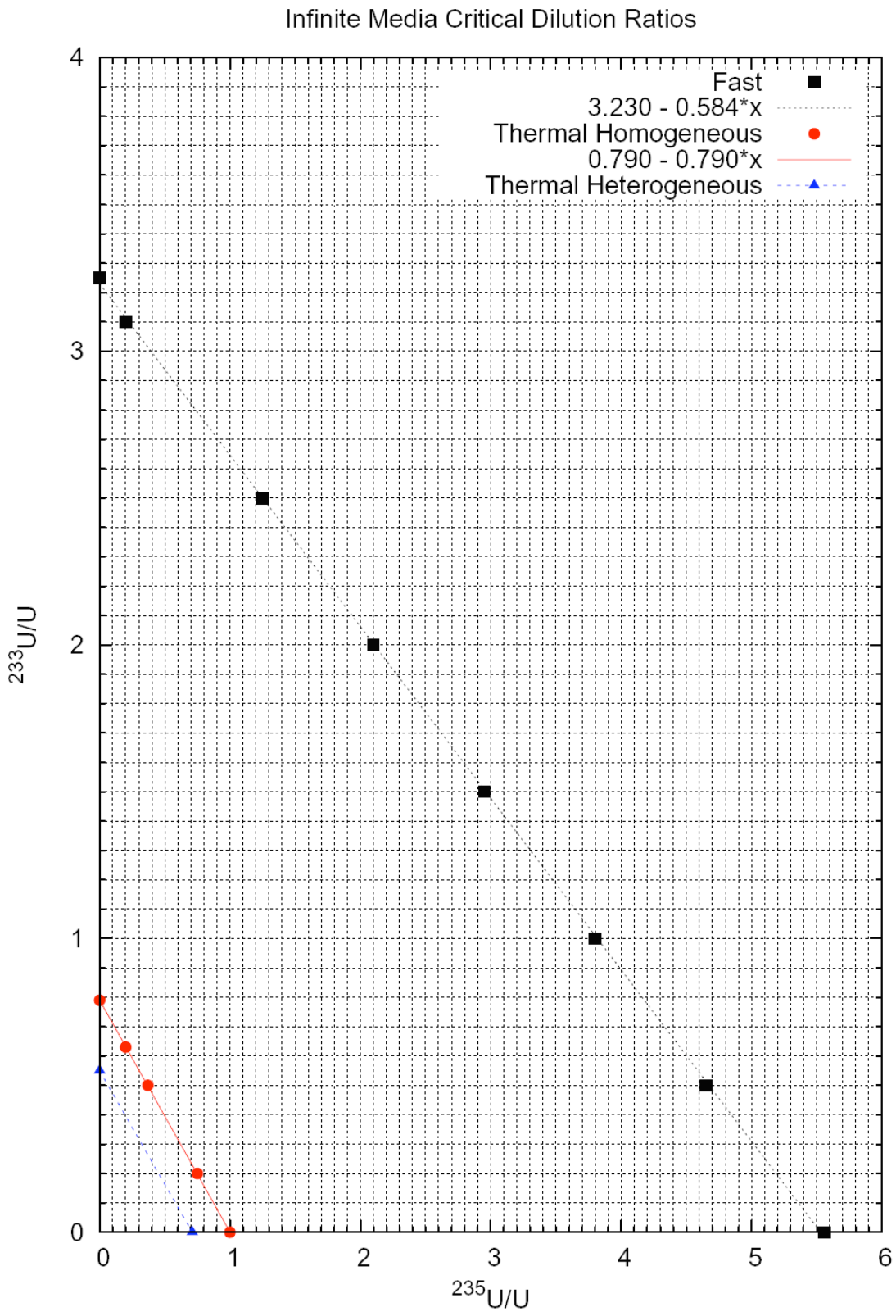
Fast dilution ratios could be calculated using integral cross sections (or spectral indices) from Scherzo 556 and compared to values determined from Big Ten or equilibrium spectra in large natural uranium assemblies. The effect of spectrum softening with increasing dilution should be discussed.

Thermal homogeneous and heterogeneous dilution ratios could be derived from integral thermal cross-sections (or 2200 m/s cross-sections with g-factors) using the four-factor formula.

## Conclusion

Fissile materials dispersed into the 'far-field' will be distributed heterogeneously among other materials (absorbers, diluents, moderators) and homogeneity cannot be assured. No discussion of heterogeneity is provided in the ORNL report.

The proposed dilution ratios (limits) in the ORNL report are derived from considerations of a homogeneous *theoretical* infinite medium at optimum moderation without absorbers. This approach while certainly conservative is sure to maximize cost.


Infinite Media Critical Dilution Ratios
Fast
3.230 - 0.584*x
Thermal Homogeneous
0.790 - 0.790*x
Thermal Heterogeneous
233U/U
235U/U
0
1
2
3
4
5
6

**Review B**

**General Comments**

1. In the absence of any practical way to burn the $^{233}U$ in a reactor or accelerator system, dilution of the $^{233}U$ with DU seems to be the most (or possibly only) feasible way to address the criticality safety aspects of disposal. The concept outlined in this document is sound although the exact values of some of the parameters may need to be revised.

2. The document was written in 1997 and references various existing documents regarding regulatory requirements, DOE decisions, etc. Have there been any significant changes in regulatory requirements, guidelines, DOE/NRC positions, etc. in the intervening years that should be incorporated in the ORNL document?

3. The calculations in the document were performed in 1996 using SCALE with the 238-energy group ENDF/B-V library. This library is somewhat dated in the spring of 2007. ENDF/B-VII was released recently. It would be desirable to check at least a few of the limiting cases in Appendix A with the most recent release of the nuclear data and with continuous energy calculations to examine the effects of the dated cross section library and of multigroup approximations.

4. As a general point, the document refers to moderation by water or hydrogen in several places. Any scattering material provides some degree of moderation. Some materials are better moderators than others, but any material that provides scattering without significant capture provides some moderation. Hydrogen and water are the standards because a) energy loss is greatest in hydrogen scattering and b) water is widely available as a scattering medium. In fact, beryllium, BeO, heavy water and graphite may be better moderators and reflectors than water because Be, O, C, and $^{2}H$ have much lower capture cross sections than $^{1}H$. Silicon also has a low capture cross section, so silicon dioxide may be comparable to water with respect to moderation in an effectively infinite medium. What really matters in determining $k_{\infty}$ is total neutron production due to (n,f), (n,2n), and (n,3n) reactions relative to total neutron loss due to the various absorption reactions. Silicon dioxide may be a very effective moderator in this case if neutron losses due to the various capture reactions are sufficiently low. This may also be true of graphite, other carbon compounds, some plastics, etc.

5. The analysis is based on homogeneous mixtures of uranium, $SiO_2$ and water. Is it possible that a heterogeneous configuration could be more reactive? After all, heterogeneity is required to make a reactor fueled with very low enrichment uranium critical. In a repository setting, material that is dissolved and transported out of the waste containers will accumulate in small cracks in the rock. A homogeneous mixture of uranium, water and rock is a first approximation, but the actual configuration is likely to consist of rock with uranium and water dispersed in small cracks of various sizes. It might be worthwhile to compare homogeneous calculations with heterogeneous calculations for alternating layers of materials to ascertain whether the homogeneous configuration is always the limiting case.

**Specific Comments**

1. Section 1.1, p. 1, para. 4 - $^{233}U$ has a smaller critical mass than $^{239}Pu$ in some energy ranges but not universally. For example, the single parameter limits for $^{239}Pu$ in ANSI/ANS-8.1-1998 are lower than those for $^{233}U$.

2. Section 3.3.2.2, p. 11, para. 3 – This paragraph suggests that it may not always be necessary that the DU be isotopically mixed with the enriched uranium in the waste package and that it is only necessary that the uranium be isotopically mixed when it is transported from the waste package. Can this be guaranteed under all credible conditions if the DU is separate from the enriched uranium in the package? Are there realistic conditions under which separated, i.e., non-mixed, DU and enriched uranium in a package could dissolve and transport at different rates or to different locations?

3. Section 3.3.2.3, p. 13, para. 3 and 4 – For the scattering materials, it is just conservative to assume that they remain. Restricting the analysis to a homogeneous mixture of uranium, water and silicon dioxide assumes that no more effective scattering/moderating material is present. Beryllium and carbon may be better moderators than water under some conditions because of their very low capture cross sections. Are there any minerals present that may contain significant amounts of carbon, e.g., in the form of carbonates? Limestone, for example, is calcium carbonate.

4. The first paragraph in Section 4.1 and the appendix both refer to the commonly accepted concept that two or more mixtures of optimally water-moderated, subcritical infinite-medium fissile materials may be homogeneously combined and remain subcritical if the composition of the materials remains homogeneous. Will this apply to or envelope the cases of optimal moderation by a water-silicon dioxide mixture? A number of cases in Table A.1 appear to have very similar values of $k_{eff}$ with significantly different mixtures of water and silicon dioxide. If uncertainties in nuclear data and processing are factored in, it may be the case that the limiting configuration actually occurs for some intermediate mixture of water and silicon dioxide rather than with 100% water. The mixture concept seems reasonable in the present case, but the discussion in Section 4.1 and/or the appendix might need to be expanded to address optimal moderation in the water-silicon dioxide mixtures.

5. Section 4.1 restricts materials to $SiO_2$, water, DU, $^{233}U$, $^{235}U$ and $^{238}U$. Are there any other minerals with better scattering/moderating properties (and lower capture cross sections) than $SiO_2$ that should be considered? Also, are there other materials in the package that could degrade and that may be good moderators with low capture cross sections?

6. Section 4.1, p. 17 states that "the limiting subcritical enrichment for $^{235}U$ (Paxton and Pruvost, July 1977) for optimally moderated homogeneous aqueous systems is well-defined to be 1 wt% $^{235}U$." Is this also true for $SiO_2$ or $SiO_2$ – water mixtures?

7. Is there any significant amount of $^{232}U$ in the $^{233}U$ waste?

8. Sections 4.3 and A.8 both exclude compounds of beryllium and deuterium. Certainly graphite and carbon compounds should be added to that list. There may be other materials that should also be added.

9. Section 4.3, p. 18, para. 1 – The last sentence states:

"Though other scattering or absorbing nuclides may be present in a mixture, their effects have not been accounted for in the reduction of required DU mass for dilution of $^{233}U$ and enriched uranium."

It is conservative to neglect absorbers. The effect of neglecting scattering nuclides depends on whether they may be better scatterers and moderators than silicon dioxide and water. Graphite, carbon compounds and some plastics may be in this category.

10. It may be implicit in the procedure, but it may be necessary to match the physical form of the DU to the physical form of the $^{233}U$, i.e., metal, $UO_2$, $U_3O_8$, etc., to ensure adequate mixing and dilution in the long term. This could add to the cost or complexity for small amounts of odd compounds.

11. Section 5, p. 19 – The conclusions assume that the limiting case is provided by an optimally-moderated homogeneous mixture of $^{233}U$. This again raises the question of whether the limiting case might be provided by some mineral other than $SiO_2$ or by some particular mixture of $SiO_2$ and water. There are a number of mixtures in the appendix that have very similar values of $k_{eff}$ and very different water volume fractions. Also, the validity of the statement that the optimally-moderated homogeneous $^{233}U$ case compensates for uncertain nuclear parameters for dry (less water-moderated) mixtures depends on the nature of the uncertainty.

12. Page A-4, eq. A.2 – This relationship may be valid for unmoderated metal, but it may not apply to a configuration that may degrade in the repository environment.

13. The cited validation reference dates from 1986 and pre-dates the ICSBEP Handbook. On p. A-5 the report notes that no benchmarks of homogeneous mixtures of uranium, water and silicon dioxide exist and raises the issue of the adequacy of silicon cross sections. ENDF/B-VII has isotopic silicon. Also, the September 2006 edition of the ICSBEP Handbook has benchmarks for mixtures of uranium with silicon dioxide and polyethylene, for uranium with sand reflectors, for $^{233}U$ solutions and possibly for other relevant configurations.

14. Section A.3, para. 2 states that silicon dioxide and water were selected as the most restrictive materials. Again, are there any other minerals with significant carbon or beryllium that may be present?

15. Table A.1 shows a number of combinations of $^{233}U$ dilution, silicon dioxide concentration and water concentration relative to $^{233}U$. It would be of value to add a table comparing 100% water versus 100% silicon dioxide over the relevant range of $^{233}U$ concentrations for the proposed dilution (or dilution range) to verify that water does provide the limiting case.

16. The report imposes the restriction that the atom ratio ($^{234}U + ^{236}U)/^{235}U$ is less than one. Is this always true for a DU/$^{233}U$ mixture? How much $^{234}U$ is present in $^{233}U$? Likely small but some values could be noted.

**Review C**

**General Comments**

1. The authors did a credible job of deriving dilution requirements for the specific scenarios they considered.

2. The dilution justification seems valid and would be a good approach in general for processing this material.

**Specific Comments**

1. The report says that $^{233}U$ not only has a smaller critical mass than $^{235}U$ or $^{239}Pu$ but also has "other fissile properties that are significantly different." It never spells out what the latter characteristics are or how they might impact this analysis.

2. The major formula presented in the executive summary (E.1) is confusing. "Total uranium" is enriched uranium plus $^{233}U$ but does not include depleted uranium. The terminology could be clearer.

3. Are the process options discussed on p. 6 still relevant ten years later? It is not clear if the scenario presented in 3.2.4 (DWPF) is being considered as an alternative to DU dilution.

4. The logic in 3.3.2.3 is not entirely clear. The assertion is made that H, O and Si are always found in uranium ore deposits. The conclusion then is that criticality in disposal sites can be prevented by considering the most reactive system of U, $SiO_2$ and $H_2O$. This may indeed be true, but the implied connection between the materials associated with uranium in ore deposits and the materials associated with uranium in disposal sites is not clear.

5. The selected subcritical value for $k_\infty$ was 0.95. There was not much discussion of the basis for selecting this value. In fact p. A-7 states that the value "is not fully justified."

6. Section 4.3 indicates that moderators such as C, Be and D were not considered. Perhaps this should be highlighted elsewhere. The concern is whether a more optimally-reactive mixture exists (even if it is very unlikely in nature) than water plus uranium.

7. The cross section library used in the SCALE analyses was based on ENDF/B-V. These data were rather dated when the report was written. They are more dated now.

8. The authors note that "no benchmarks of homogeneous uranium metal, water, and $SiO_2$ mixtures exist." This seems to be a relatively important point. It should not be buried in the Appendix.